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Lifetime Measurements of Highly Charged Ions

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Abstract

How long does an atom "live" in an excited state, when due to conservation rules and symmetry principles the "normal" electric dipole decay to the ground state is forbidden? Then spin-changing "intercombination" transitions and electric-dipole "forbidden" decays occur that are important for the diagnostics of low-density solar coronal and terrestrial plasmas. Regular "allowed" and intercombination transitions in highly charged ions (picosecond and nanosecond lifetime range) have previously been measured with straight, foil-excited, fast ion beams. For ions with only one or two electrons in the valence shell, very long sections of the isoelectronic sequence have been covered this way, yielding experimental tests of theoretical predictions of transition rates that are good to a few percent. Recently the situation has become much better than this for lifetimes in the millisecond range. Here the technique of optical observation at a heavy-ion storage ring permits atomic lifetime measurements on intercombination and forbidden transitions with a precision that often is better than 1%, which is clearly surpassing the precision range of typical conventional ion trap data. The electron beam ion trap (EBIT) exhibits a similar precision in the "X-ray" range, and it has reached uncertainties of a few percent in some measurements of optical transitions.

1. Introduction

Lifetime measurements of ions have been a major topic at quite a number of scientific meetings, in particular in the hey-days of beam-foil spectroscopy. Quite a number of introductions and reviews have been written on the subject, of which I will not quote more than a few [1–5]. I'd rather concentrate on more recent developments like that offspring of beam-foil spectroscopy, lifetime studies using a heavy-ion storage ring, that first saw the ASOS conference light at Meudon, and on the uses of the electron beam ion trap (EBIT). "Ions" in the title of this talk allow me to skip over the excellent lifetime work on fast atoms and rare-gas jets done at Kaiserslautern [6], except for mentioning that some of the storage ring results have similarly small uncertainties. "Highly charged ions" permit me not to go into Eric Pinnington's excellent ion-beam laser work on singly charged ions of astrophysical interest [7,8]. Of course, his now dissolved laboratory was not the only one, and precision lifetime work on ions in low charge states continues at various universities, like by Jim Lawler's group at the University of Wisconsin, by the Richard Holt/David Rosner group at the University of Western Ontario, or at Sune Svanberg's laboratory at Lund, naming just a few. (I know, there are plenty of others not mentioned – be assured that you are in good company.) I have even done some work on low-charge state ions at the ion storage ring myself (which therefore must be included), there is excellent work on singly charged positive ions at the Stockholm CRYRING and on negative ions at the Aarhus ASTRID and ELISA storage rings. My selection is based on techniques that are suitable for lifetime measurements on highly charged ions, and will put emphasis on precision measurements and on ions of astrophysical interest. For discussions, reviews and further literature listings I recommend Indrek Martinson's series of Comments on the beam-foil method [9–12] and venture to refer to my own recent review on the lifetime measurements done with ion traps [13].

2. Some basic atomic physics points

The study of atoms has many aspects. Leaving aside the response of atoms to outside influences (collisions, external fields), we pretend that there is the atom *per se* that ought to be understood from its emission spectrum, that is from the light emitted after whatever cause has excited the atom. The fact that the light spectrum (in emission and absorption) has individual features points to the level structure of the atom. But the observation that not all level differences appear as lines in the spectrum, and that existing lines have characteristically different relative intensities, points towards selection rules which reflect symmetries of the atom and its interaction with the radiation field [14].

There are transitions (between levels k and i) that take place with a high transition probability A_{ki}, and others that are less probable by orders of magnitude. An inspection of the symmetries involved reveals that the most probable transitions of the atom correspond to electric dipole (E1) transitions which connect levels of opposite parity (Fig. 1). A series expansion of the radiation field has magnetic dipole (M1) and electric quadrupole (E2) components next (and so on, M2, E3 $\cdots$), but they usually are of much lower transition probability (and connect to other levels, of the same parity), so that they do not matter as long as an El transition is allowed. However, sometimes this decay path is blocked for an atom, and there may only be lower levels of the same parity, for example the other fine structure levels of the same term (level multiplet). Also, the higher-multipole transitions scale faster with the nuclear charge along an isoelectronic sequence (ions of different elements, but with a constant number of electrons). Thus in highly charged ions the higher-multipole order,

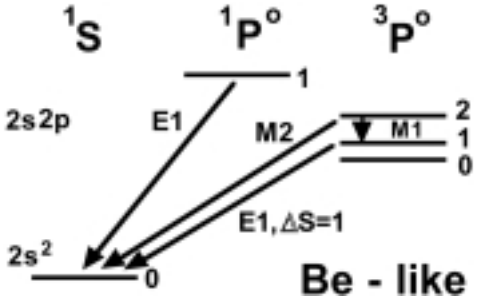

Fig. 1. Level scheme of Bc-like ions, indicating various transition types.

electric-dipole "forbidden" transitions become more prominent. With the right light source, be it the solar corona or an electron beam ion trap, one can even find spectra that are dominated by electric-dipole forbidden lines.

These increases have long ago been systematized by Hylleraas [15] and depend on two factors. One is a dependence on the transition energy (which itself has a characteristic trend, like the Z^2 dependence of levels of different n in Bohr's model of the hydrogen atom which describes atomic shell separations, or the linear dependence on Z of energy differences within a given shell of a multi-electron system ($\Delta n = 0$)). The other is the "atomic structure" part, be it expressed as line strength, oscillator strength, multiplet mixing parameter, hyperfine interaction or the like.

Typical level lifetimes (the inverse of the sum of all decay probabilities from a given level) dominated by E1 transitions are in the nanosecond range for neutral atoms and become too small (sub-picosecond range) to measure with precision for only moderately highly charged ions (E1 $\Delta n \neq 0$ transition rates scaling with Z^4, with n the principal quantum number). However, transitions within a given electron shell scale in both, transition energy and rate, only linearly with Z and may therefore be followed along very long sections of an isoelectronic sequence. Transitions that require a spin change (E1 $\Delta S = 1$, that is, *intercombination* transitions connecting singlet and triplet, or doublet and quartet, term systems) or that are E1-forbidden, have a very small probability in neutral atoms and may become measurable only in multiply charged ions, because of the steep increase of their transition probabilities with the nuclear charge Z.

Important for the present discussion are just a few general points:

- A decay mode of interest may be accessible to study only in a few ions, where it is not dominated by some other decay or competing collision processes.
- A transition of interest may be accessible to a given instrumental technique only in a particular range of ion charge states, because of its transition energy (wavelength range) or lifetime. For example, fine structure intervals scale with Z^4, and thus the wavelengths of the associated forbidden transitions vary drastically along the isoelectronic sequence. This is a problem for theory, too, as calculations are still struggling with *ab initio* predictions of such fine structure intervals in complex ions to better than, say, 10%.

Intercombination and forbidden transitions in low-charge state ions are of great interest in astrophysics, because their observation reaches larger optical depths than the observation of allowed transitions. In higher charge state ions, intercombination and forbidden transitions are of value for the diagnostics of solar and terrestrial plasmas [16–19]. Forbidden transitions in singly charged ions are also intended for use in frequency standards, because the low transition probabilities result in small level and line widths. However, only very recently any precise measurements (accuracies in the single-percent range or better) have been achieved on intercombination or forbidden transition rates in any ion.

The quest for a detailed understanding of atomic structure is certainly helped by independent checks on the quality of the wavefunctions used. The variational principle identified by Ritz only applies to energy levels (any imperfect wavefunction will yield energy levels higher than the true ones). E1-transition probabilities, in contrast, depend differently on the radial wavefunctions. The latter need not be orthogonal to each other and are nowadays optimized separately for initial and final states of a given transition [20]. The calculation of forbidden transition rates is very sensitive to the wave function composition used. In the absence of a guiding principle from theory, however, only experiment can provide precise tests of atomic lifetime calculations.

3. Experimental techniques

For several decades, beam-foil spectroscopy (Fig. 2) has been the work-horse for lifetime measurements in any element and ionization stage. This is because any charge state of any element can be reached - the highly charged ions of very heavy elements requiring rather large ion accelerators - and because the technique offers inherent time resolution. One only needs to vary the distance between the exciter foil and the line of sight of the detection system, and a wavelength selector (filter or spectrometer, preferably with a multi-channel detector). Easily achieved displacements from a few μm to, say, a meter, translate into observation times after excitation that range from about a single picosecond to dozens of nanoseconds. This range is sufficient to follow the resonance transitions of Na-like ions from the neutral atom up to Au^{68+} [21–23]. With selective excitation by laser light, high precision lifetime values have been obtained on neutral atoms (like Li and Na) and singly charged ions (like Mg^+). For multiply charged ions the energy intervals are too large to be bridged by laser light, and the precision then reached after non-selective excitation is much lower. In spite of correlated analyses that take care of cascade influences, the typical beam-foil lifetime precision is not better than a few percent (there are a few good and many bad exceptions). However, data along isoelectronic sequences can be analysed together, reducing the uncertainty of the average.

However, the consistency of the emerging data sets may depend on hidden parameters: In one of the most recent papers on lifetime measurements of the $3p_{3/2}$ level in Na-like ions [23], the experimental data are compared with theory. Data for 12 ions, from Si^{3+} to Xe^{43+}, are shown that are quoted from 10 papers with a total of 67 authors. Of these, some appear repeatedly, so that only 40 different individuals are named. While the work was done at accelerators of all sizes (at Argonne, Bochum, GSI Darmstadt, RIKEN Tokyo, and Toledo), it turns out that just four people among them

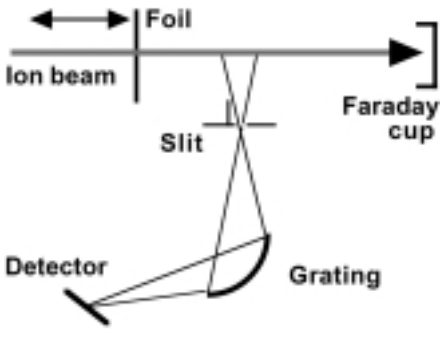

Fig. 2. Schematics of beam-foil spectroscopy.

have participated in all of these studies (L.J. Curtis, L. Engström, R. Hutton, E. Träbert). Therefore the question arises whether the consistency is a product of the technique or rather of its practitioners. After a quarter of a century in this business, I dare state that not everybody in the field seems to have appreciated the physical situation of the beam-foil measurements nor realized the intricacies of the atomic structure that underlies the decay curves obtained. A lot of wildly scattered lifetime data, in particular in the early years, thus gave beam-foil an undeservedly poor reputation. With some atomic physics insight, however, reliable lifetimes can be gained, but that brings in a human factor and personal judgement. There are very few cases in which valid atomic lifetimes can be determined (in the sense of signal curves being evaluated) almost completely without human interference; for example, after selective excitation (by laser), or on light ions in a storage ring, where a single metastable level may be contributing (see below).

The only isoelectronic sequences that have been studied for level lifetimes over as wide a range as the Na sequence are those that feature transitions within a given electronic shell ($\Delta n = 0$), because for such E1 transitions both the energy interval and the transition rate scale linearly with the nuclear charge Z or the ion core charge. Other types of transitions scale so much more steeply with Z that only part of an isoelectronic sequence is in any lifetime range that is accessible by present techniques. For example, experimental problems so far have largely precluded atomic lifetime measurements in the microsecond range that is typical for intercombination transitions in moderately charged rather light ions [24].

With ordinary E1 decays blocked and only other decay modes available, however, there are levels that in low-charge ions may have lifetimes that are much, much longer than those mentioned so far. Quite a number of them are as high as milliseconds or seconds. Of course, in order to see the radiative decays of such levels, the collision frequency must be lower than the inverse level lifetime, and that corresponds to environments of, say, 10^{10} particles per cubic centimeter. Such densities are typical for coronal plasmas, and hence it is not surprising that the millisecond- and second-lifetimes are of astrophysical importance. On Earth, their study requires ultrahigh vacuum (UHV).

However, this is not all: Fast ion beams of 2 MeV/nucleon travel at a velocity of 2 cm/ns. Lifetimes of 1 ms thus correspond to 20 km of ion travel. One way of avoiding the cost of such extended UHV systems is the formation of the beam tube into a storage ring, in which the ions can circulate, passing the same detector over and over again (Fig. 3). As with yarn on a spool, the ion beam in the ring needs to be slightly displaced after a turn in order to accept the next turn. This ''stacking'' technique permits the accumulation of a stored ion beam over about 30 turns (at TSR Heidelberg) before the phase space of the ring is filled. It is possible to cool the stored ion beam by superimposing a ''cold'' (monoenergetic) electron beam and thus to increase its phase space density. However, such cooling usually takes more time than the lifetime of the excited levels of interest. Consequently the choice of options is limited to lifetime experiments that depend either on the initial excitation process (in the ion source or the injector) and do not employ cooling, or which use cooling and subsequent electron capture (in a gas jet target or in the electron cooler). If the excitation interval is very small (as is the case for hyperfine structure in the ground state), laser excitation may be possible [25–27]. Laser excitation and level population manipulation are also used at the Stockholm storage ring CRYRING, where singly charged ions are accelerated in the ring, that is after injection. Of particular interest in the astrophysical context of this conference is their lifetime measurement of four important (out of 62 in total) low-lying levels of Fe^+ [28].

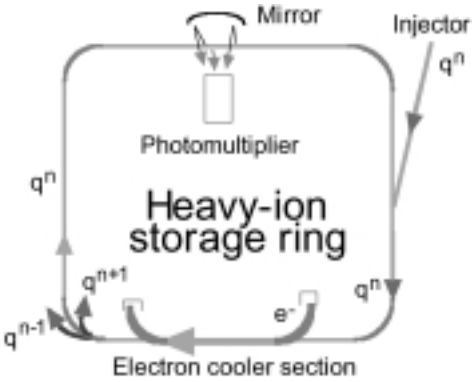

Fig. 3. Schematics of a heavy-ion storage ring with particle detection (bottom) and photon detection (top).

Storage rings come in many sizes and with different accelerators for injection. Consequently the physical systems that can be studied differ. This is the reason for the apparent specializations in atomic lifetime measurements at CRYRING (singly charged heavy ions), ASTRID/ELISA (negative ions), ExperimentSpeicherRing ESR (very highly charged few-electron ions) and TestSpeicherRing TSR (low to moderate charge state ions). Technical advantages of all such storage rings compared to conventional ion trap measurements lie in the spatial separation of ion production and selection of a single iosotope and charge state on one hand (in the injector and beam transport system), and of storage (under UHV conditions at MeV energies) and detection on the other, which can be optimized separately. The result are reductions of the experimental errors often by more than an order of magnitude, and the outright removal of some sources of systematic error that haunt other techniques.

However, not everybody has a storage ring nearby, and cheaper devices might be sufficient for some tasks that require ion trapping for atomic lifetime studies. The basic working principles of the conventional electrostatic (Kingdon), magnetic (Penning) and radiofrequency (Paul) traps have been outlined repeatedly elsewhere, as have those of electron-beam ion traps and ion storage rings, and they need not be reiterated here. New trap types based on electrostatic mirrors have been developed in Israel [33–35] and Stockholm [36], but those devices so far are used with low-charge state ions only.

Lifetime work with all ion traps has to ascertain the influence of collisions as a competing mode of destruction of the ion population. Obviously this problem is more severe for classical traps that operate in the pressure range near 10^{-8} mbar than in the latest UHV Kingdon traps ($p \approx 5 \cdot 10^{-10}$ mbar) [37], the electron beam ion trap with its cold surfaces ($p < 10^{-10}$ mbar) or the heavy-ion storage ring ($p \approx 10^{-11}$ mbar). Moreover, the heavy-ion storage ring offers on-line monitoring of the single stored ion species via the ion current. EBIT ion storage can be controlled via ion

cyclotron frequency measurements [38] or via X-ray monitoring of charge exchange processes. At the Kingdon trap, the ions that are ejected at switching off the trap voltages are being used to determine the trapping properties [39].

Correcting measured (apparent) lifetimes for the ion storage time always results in a longer "true" lifetime value. There are a few cases for which comparison data are available from an ECR ion source plus Kingdon trap, EBIT and the heavy-ion storage ring. The latter device has the longest ion storage times and hence the least need for correction. Moreover, the storage time measurement on the single-charge ion beam in the storage ring is the most direct and the cleanest, and thus the procedure with the smallest uncertainty. For atomic lifetimes in the sub-ms range, storage time corrections are generally very small, and results from different trap types agree with the same trend [40,41]. For longer lifetimes, the storage-time corrections are more critical, and here the storage ring provides the best conditions.

Photon detection profits from a large solid angle of detection which captures as much of the available light as possible. However, ion trap geometries impose certain limits. While small ion traps have been built for single ions, other traps (and ion clouds) have sizes in the cm-range, while heavy-ion storage rings for atomic physics have circumferences of 10 to 100 m. In the latter cases, the extended light source does no longer permit to observe more than a small fraction of the ion cloud by a single detector. With fast ion beams, Doppler shifts and spreads add complications, advantages and disadvantages, to a given design. In contrast, Doppler shifts are practically absent from spectra recorded at the electron-beam ion trap, because EBIT offers spectroscopic access to highly charged ions that are practically at rest. The basic idea exploits the same collisional production of highly charged ions as the fast-ion beam-foil work, only with the roles of projectile and target interchanged. The tightly collimated electron beam also helps to confine the ion cloud radially, while slots in the innermost cold shroud as well as windows and flanges limit the viewing zone to typically 2 cm in length. In order to maximize the light use, it is advantageous to use this emission zone as the "entrance slit" of a detection system.

Atomic lifetime measurements with EBIT have been done in two very different lifetime ranges, femtoseconds (by a linewidth measurement on Ne-like Cs^{45+}, reducing the thermal motion Doppler width to below the natural line width [42]) and milliseconds. We concentrate on the latter for the astrophysically relevant measurement of forbidden decay modes. With X-ray detectors (available in sizes that make large solid angle detection possible) each signal pulse may be analyzed for its energy, and many false counts can be rejected, resulting in very clean data. The high signal-to-noise ratio of X-ray measurements makes it much easier to obtain precision lifetime data in the X-ray range than in the visible [43].

Millisecond-atomic lifetime measurements in EBITs nowadays rely on the sharp switching off of the electron beam [44,45]. The physical difference with an earlier energy-modulation technique is that in the older technique a partial space-charge compensation by the lower-energy, full-current electron beam remained, while in the new technique all further electron-ion collisions are intentionally avoided by the lack of free electrons, at the cost of a change of geometry of the trapped ion cloud that expands radially to a new equilibrium. Although the electron beam has been switched off and thus the attractive potential and space-charge compensation by the electron was removed, an EBIT in the magnetic trapping mode [44] then continues to provide trapping like any Penning trap, for many seconds.

4. Atomic systems of interest

Conventional beam-foil spectroscopy, with straight ion beams, has been so successful at ion accelerators of all sizes that most cases of high interest that are amenable to this technique apparently have been covered. Of course, this technique remains unique in its ability to produce multiply and highly excited ions of all charge states of all elements, but the high charge states of heavy ions require high-energy (in the atomic physics sense) accelerators of which not many exist. There is plenty of useful and interesting work left to do with beam-foil spectroscopy, but almost 40 years after its inception it is more difficult than ever to have it funded. Also, some of the pure spectroscopy (for any charge state of any element, although for low-lying levels only) would better be done using electron beam ion traps. For lifetime studies in the range below some 20 ns, in highly charged ions, beam-foil spectroscopy remains the only tool available.

However, there are interesting atomic systems with level lifetimes very different from those just mentioned – longer by 4 to 8 orders of magnitude, and these will be the topic for the rest of my presentation. As an illustration, a selection of the presently available ion trap lifetime data on forbidden transitions is given in Table I. Some of the work employs a straightforward extension of the beam-foil idea, by feeding a foil-excited ion beam into a heavy-ion storage ring. The lifetime measurements on those long-lived levels, however, have turned out to be not just "more of the same", albeit on systems that are of interest for various branches and applications of physics. Instead, they have reached an unprecedented level of precision that even exceeds the needs of the various applications, and that severely tests calculational approaches.

4.1. *He-like ions*

Hydrogenlike (single-electron) ions are considered to be so well calculable that lifetime measurements are not expected to be able to test theory in a serious way. In the last few years, extreme calculational precision has also been reached for He-like (two-electron) ions. In fact, one of the longest isoelectronic sequences ever covered by lifetime studies is the $1s^2\ {}^1S_0$ – $ls2s\ {}^3S_1$ transition in He-like ions. The rate of this transition has been studied from neutral He (by laser absorption, the lifetime exceeding 1 h) to Xe^{52+} (by beam-foil spectroscopy, picosecond lifetime), that is for lifetimes that span 15 orders of magnitude (see [46]). In between these extremes, various ion traps, a heavy-ion storage ring, an electron-beam ion trap, and a slow (recoil) ion beam have been employed. Eight years ago, only two heroic, but not precise, measurements existed for He and Li, and (mostly) beam-foil measurements covered a fair number of elements from $Z = 16$ (S) to $Z = 54$ (Xe). None of the measurements was more precise than about 5%. A very precise, largely non-relativistic calculation [47] with a leading-term relativistic correction [48] was a good guide, but was expected to be possibly insufficient at high Z. The relativistic calculations

of the time, however, yielded rather diverse predictions in the low-Z range.

Since 1994, heavy-ion storage ring data, obtained by populating the level of interest via dielectronic recombination in the electron cooler section of the Heidelberg storage ring TSR, suddenly changed the situation. They reached a precision as good as 0.2% [49,50]. This must have inspired new efforts on the theory side bearing fruit in extensive relativistic calculations [51] that at long last (26 years later!) matched Drake's low-Z results [47] and are expected to be valid all along the isoelectronic sequence (Fig. 4). For a comparison of these two sets of calculations that both employ near-perfect wavefunctions, some Livermore EBIT results [40,52] are the most sensitive ones on the market. Their precision (for $Z = 8$ and $Z = 10$) of just below 0.5% is quite sufficient to reject most of the earlier calculations. Unfortunately – from the experimenters' perspective – the two good calculations differ by only 0.1% in this range (and by 0.2% for $Z \geq 18$), challenging experiment to come up with another order-of-magnitude improvement in precision – which is not yet in sight. For the moment, the agreement of precise heavy-ion storage ring and EBIT lifetime measurements with those impressively accurate calculations is exploited as a reference for the quality and validity of the experimental techniques employed.

4.2. *Be-like to Mg-like ions*

Beam-foil spectroscopy has been employed to measure intercombination transition rates in He-like ions (mostly $n = 2$ levels), for the intercombination transition decays of the $n = 3$ levels in Be-like ions (see [12] and refs. therein), and for the $n = 2$ resonance level decays in Be-like ions. However, the experimental lifetime data rarely reach a precision of better than 5%. For the resonance line transition probability in such Be-like ions, theoreticians are confident that their large-scale calculations are good to a few parts in ten thousand, which seems out of reach for present experimental techniques. The situation is very different for the ground-state intercombination transition in such ions. For several cases the experimental lifetime precision reached is excellent, while theory is facing serious obstacles. These relate to the fact that either, in a non-relativistic approach, the Breit interaction, a small entity, needs to be calculated with precision, or, in a relativistic setting, the small difference between two large wave function components.

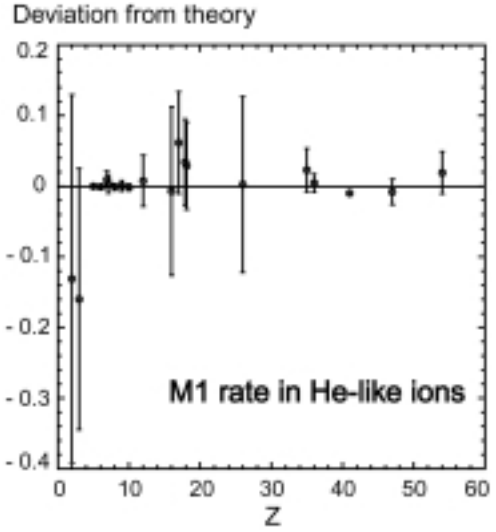

Fig. 4. Iso-electronic trend of the transition rate data for the $1s^2\ ^1S_0$ - $1s2s\ ^3S_1$ magnetic dipole (MI) transition in He-like ions. Data for $Z = 3$ to 12 are from the heavy-ion storage ring TSR and from the Livermore electron-beam ion trap (EBIT), respectively. All high-Z data are from fast ion beams, with the exception of one datum for $Z = 18$ which is from a slow ion beam. The (selected) experimental data have been scaled by reference to the fully relativistic calculation by Johnson *et al.* [51] (Horizontal line).

Early calculations of the rate of the $2s^2\ ^1S_o$ - $2s2p\ ^3P^o_1$ intercombination transition in four-electron ions (Be iso-electronic sequence) scattered markedly, and the uncertainties of experimental (mostly beam-foil) lifetime data on highly-charged members of the Be iso-electronic sequence were at the 10% level and worse. For the Be-like ion of highest astrophysical interest, C^{2+}, predictions and radiofrequency ion trap data scattered by about 20% around the mean. Then calculations from Belfast and Nashville came along, with results near the sample mean and clearly contradicting the published experimental evidence from radio-frequency ion traps. The first heavy-ion storage ring data on C^{2+} [53] and B^+ [54] with their sub-1% uncertainties confirmed this theoretical claim forcefully.

The calculations so far had barely reached an uncertainty estimate of about 1% [55,56]. Very recently, however, a relativistic configuration-interaction calculation with about 200 000 configurations claimed a precision of only 0.5% [57] - and stated a disagreement by 1.5% with the 0.14% storage ring result, that is clearly beyond the mutual error bars. The strong point of the new calculation is that the authors found a way to obtain a rather good agreement of the results from length and velocity forms of the transition operator, by taking negative-energy states into account. This is a marked improvement over the previous relativistic calculations of this system (and a very interesting basic physics finding in itself - negative energy states changing a particular physical prediction by a factor of two, ending up close to where other ways of prediction went previously). Earlier on Hibbert stated [58] that in case of agreement between theory and experiment possibly neither is right. With the fresh disagreement of the latest precisely measured and calculated results on the intercombination transition rate in C III we now have the (much clearer?) case in which we know that one result may be less right than the other – but which one is it? The challenge is back!

Unfortunately, there are no precise lifetime measurements available yet for Be-like ions heavier than C, which would also help identifying any possibly overlooked systematic error in the measurement on C^{2+}. There are plans (depending on the funding and manpower) for a detector inside the heavy-ion storage ring vessel that should eventually permit to obtain equivalent data on N^{3+} and O^{4+}. However, the same calculation that describes well the low-Z ions of the sequence [59] (Fig. 5), seamlessly matches a fully relativistic calculation for high nuclear charges Z [60]. It is also compatible with all beam-foil data in the medium-Z region (reaching from Fe^{22+} to Xe^{50+}), and one may safely assume that it is more precise and reliable than all the experimental data on the Be sequence beyond $Z = 6$ (C) that are on the market right now.

The Mg iso-electronic sequence features the same valence shell level structure as the Be sequence as far as levels are occupied, and it ought to cause comparable problems to

calculate the $ns^2\ ^1S_0$ - $nsnp\ ^3P^o_1$ intercombination transition rate of electrons outside a core of closed shells, for $n = 3$ as for $n = 2$. However, in all such progressions one finds that the heavier ions have higher intercombination transition rates than the lighter ones. This is likely caused by the penetrating s and p electron orbits that take little notice of inner electrons that may have larger angular momenta. Hence the low-angular momentum valence electrons experience not the same core charge as indicated by the ion charge (plus one), but, with increasing principal number of the valence shell, a (relatively) less and less well screened nuclear charge. Incidentally, the wavelengths of the $2s^2\ ^1S_0$ - $2s2p\ ^3P^o_1$ transition in B^+ and of the $3s^2\ ^1S_0$ - $3s3p\ ^3P^o_1$ transition in Al^+ are almost identical, as was exploited in a storage ring experiment [54]. The measured lifetimes, 100 ms and 300 μs, however, differ considerably, and the steep scaling precludes further investigations by the storage ring technique of the same transition in heavier two-electron ions.

Until very recently, the theoretical coverage of the Mg sequence was unsatisfactory. Calculations existed for either low or low-to-medium, or medium-to-high charge states, and no obvious connections between predicted data regions existed, which discouraged simple interpolations in order to bridge the gaps. A set of wide-range investigations and calculations by Zou and Froese Fischer [61] now spans the full sequence with predictions based on the same calculational approach (although it does not provide numbers for all elements of interest). The new predictions basically agree with the older ones within their range of scatter.

Thus, curiously, a discrepancy with a subset of the experimental lifetime determinations on the intercombination transition $3s^2\ ^1S_0$ - $3s3p\ ^3P^o_1$ persists. Up to about $Z = 35$, beam-foil lifetime data from Bochum [4] and from Argonne [62] are compatible with prediction. High-Z data for Xe ($Z = 54$) and Au ($Z = 79$) [63] carry larger uncertainties and suffer from insufficient spectral resolution and thus are less meaningful. However, measurements at RIKEN (not all of which have been published) mysteriously, but consistently, deviate from the predicted trend by up to some 30%, in the range $Z = 26$ to almost $Z = 50$ [64,65]. While geometry effects have been taken care of by employing a traveling screened Faraday cup and similar auxiliary devices, a possible explanation might be seen in a phenomenon reported by Zou *et al*. [66]. According to that work, lifetime measurements performed well below the optimum ion beam energy for the production of the desired charge state ions may suffer from satellite line contamination, and the additional decay channels available to core-excited ions (autoionization) would then lead to apparent lifetimes that are much shorter than the true ones. While this hypothesis might fit the size and sign of the observed discrepancy, it has not yet been verified on the ionic systems of interest. However, the beam-foil experimental set-up at RIKEN has been dismantled meanwhile, so that such systematic tests would have to be done elsewhere.

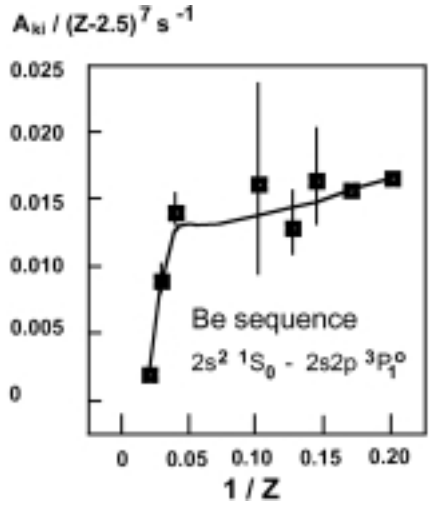

Fig. 5. Theoretical and experimental transition rate data (scaled) on the $2s^2\ ^1S_0$ - $2s2p\ ^3P^o_1$ transition in Be-like ions. For references, see [59].

4.3. *Be-, B- and F-like ions*

Both B- and F-like ions have a $^2P^o$ ground state (only the $J = 1/2, 3/2$ level sequence differs), giving rise to a single fine structure transition that is prominent in solar spectra and which thus provided an early incentive for systematizations. The same prominence makes these ions attractive for lifetime measurements. These have been tried using the combination of ECR ion source and Kingdon trap [39,67], electron beam ion traps at NIST and LLNL [68–70], and the Heidelberg ion storage ring [54,71]. In an isoelectronic comparison, the results (Table I, Fig. 6a,b) from the heavy-ion storage ring and the recent Livermore EBIT data fit a consistent isoelectronic trend that happens to coincide neatly with predictions (all calculations lie within about 2% of the reference shown, although intrinsic theoretical uncertainty estimates run up to 10 or even 20%). The Kingdon trap and NIST EBIT results fall notably short.

There also is an M1 transition in the first excited configuration of Be-like ions (Fig. 6c). Measurements at two EBITs [69,70,72], using different techniques, arrived at results that are similarly compatible with representative predictions [73,74], whereas a Kingdon trap result [39] differs from those by much more than the error estimate.

4.4. *Ions with more than two electrons*

Each additional electron in the valence shell increases the complexity of the spectra and of the calculations needed for interpretation. Ions of the iron group that feature partly-filled 3p shells are prominent in the solar corona (see below). The simplest of these ions are part of the Al isoelectronic sequence, and lifetime measurements on $3s^23p\ ^2P^o$ - $3s3p^2\ ^4P$ E1 intercombination transitions have been performed on ions from Si^+ [75] through Au^{66+} [76]. However, very few experimental lifetime data are available on the particularly long-lived $3s3p^2\ ^4P_{3/2}$ level, and in the middle of the periodic table of the elements, time aforementioned RIKEN effect may well be present in the data from that laboratory [64].

In the ground complexes of these $n = 3$ shell ions (Al-like to Cl-like), the same level structure as in B-like to O-like ions is repeated. There is the additional challenge for theory to deal with another closed shell, and there is the astrophysical aspect of the appearance of such ions of iron group elements in stellar plasmas. Particularly prominent in the visible spectrum are M1 transitions within the ground configurations of these ions. This prompted a series of measurements at the University of Nevada Reno, in particular on Mn ($Z = 25$) and Fe ($Z = 26$), using an ECR ion source and a

Table I. *Selection of lifetime measurements on ions with M1/E2 transitions.*

Ion	Level	Lifetime τ	Comment/Ref.
He Sequence			
B^{3+}	$1s2s\ ^3S_1$	(149.8 ± 0.45) ms	HSR [50]
C^{4+}	$1s2s\ ^3S_1$	(20.63 ± 0.05) ms	HSR [49]
		(20.589 ± 0.042) ms	HSR [50]
N^{5+}	$1s2s\ ^3S_1$	(3.905 ± 0.05) ms	HSR [49]
		(3.92 ± 0.13) ms	EBIT [44]
O^{6+}	$1s2s\ ^3S_1$	(956 ± 5) μs	EBIT [52]
Ne^{8+}	$1s2s\ ^3S_1$	(91.7 ± 0.4) μs	EBIT [40]
Mg^{10+}	$1s2s\ ^3S_1$	(13.61 ± 0.49) μs	EBIT [90]
Be Sequence			
Ar^{14+}	$2s2p\ ^3P^o_2$	(15.0 ± 0.7) ms	EBIT [72]
		(13.4 ± 0.7) ms	EKT [77]
		(15.0 ± 0.8) ms	EBIT [69]
K^{15+}	$2s2p\ ^3P^o_2$	(7.6 ± 0.5) ms	EBIT [70]
B Sequence			
Ar^{13+}	$2s^22p\ ^2P^o_{3/2}$	(8.7 ± 0.5) ms	EBIT [68]
		(9.12 ± 0.18) ms	EKT [77]
		(9.70 ± 0.15) ms	EBIT [69]
K^{14+}	$2s^22p\ ^2P^o_{3/2}$	(4.47 ± 0.10) ms	HSR [70]
Ti^{17+}	$2s^22p\ ^2P^o_{3/2}$	(0.627 ± 0.010) ms	HSR [71]
C Sequence			
O^{2+}	$2s^22p^2\ ^1S_0$	(530 ± 25) ms	HSR [93]
F^{3+}	$2s^22p^2\ ^1S_0$	(304 ± 5) ms	HSR [93]
Si^{8+}	$2s^22p^2\ ^1D_2$	(38.3 ± 0.3) ms	HSR [91]
N Sequence			
S^{9+}	$2s^22p^3\ ^2P^o_{1/2}$	(5.20 ± 0.15) ms	HSR [93]
S^{9+}	$2s^22p^3\ ^2P^o_{3/2}$	(2.10 ± 0.06) ms	HSR [93]
O Sequence			
F^{+}	$2s^22p^4\ ^1S_0$	(423 ± 10) ms	HSR [94]
Ne^{2+}	$2s^22p^4\ ^1S_0$	(223 ± 11) ms (2 σ)	RFT [86]
		(213 ± 4) ms	HSR [83]
Si^{6+}	$2s^22p^4\ ^1D_2$	(63.6 ± 0.7) ms	HSR [91]
Ar^{10+}	$2s^22p^4\ ^3P_1$	$(14.8 \pm 1.1$–$0.48)$ ms	EKT [67]
F Sequence			
Ar^{9+}	$2s^22p^5\ ^2P^o_{1/2}$	$(8.53 \pm 0.24$–$0.17)$ ms	EKT [67]
		(8.70 ± 0.37) ms	EKT [77]
		(9.32 ± 0.12) ms	EBIT [69]
K^{10+}	$2s^22p^5\ ^2P^o_{1/2}$	(4.44 ± 0.10) ms	EBIT [70]
Sc^{12+}	$2s^22p^5\ ^2P^o_{1/2}$	(1.00 ± 0.03) ms	HSR [54]
Ti^{13+}	$2s^22p^5\ ^3P^o_{1/2}$	(0.513 ± 0.010) ms	HSR [71]
Al Sequence			
Mn^{12+}	$3s^23p\ ^2P^o_{3/2}$	(31.32 ± 1.82) ms	EKT [78]
Fe^{13+}	$3s^23p\ ^2P^o_{3/2}$	(17.52 ± 0.29) ms	EKT [37,79]
Si Sequence			
Mn^{11+}	$3s^23p^2\ ^1S_0$	(1.5 ± 0.2) ms	EKT [78]
Mn^{11+}	$3s^23p^2\ ^1D_2$	(11.16 ± 0.10) ms	EKT [78]
Fe^{12}	$3s^23p^2\ ^1D_2$	(6.93 ± 0.18) ms	EKT [37]
		(8.0 ± 0.1) ms	HSR [95]
Kr^{22+}	$3s^23p^2\ ^3P_2$	(6.82 ± 0.1) ms	EBIT [92]
P Sequence			
Ar^{3+}	$3s^23p^3\ ^2P^o_{3/2}$	$(243 \pm 73$–$79)$ ms	EKT [67]
Mn^{10+}	$3s^23p^3\ ^2P^o_{3/2}$	(3.0 ± 0.2) ms	EKT [78]
Fe^{11+}	$3s^23p^3\ ^2P^o_{3/2}$	(1.85 ± 0.24) ms	EKT [37]
		(1.70 ± 0.02) ms	HSR [95]
Mn^{10+}	$3s^23p^3\ ^2P^o_{1/2}$	(6.17 ± 0.29) ms	EKT [78]
Fe^{11+}	$3s^23p^3\ ^2P^o_{1/2}$	(4.38 ± 0.42) ms	EKT [37]
		(4.1 ± 0.12) ms	HSR [95]
Mn^{10+}	$3s^23p^3\ ^2D^o_{3/2}$	(35.1 ± 1.43) ms	EKT [78]
Fe^{11+}	$3s^23p^3\ ^2D^o_{3/2}$	(20.35 ± 1.24) ms	EKT [37]
		(18.0 ± 0.1) ms	HSR [95]
Fe^{11+}	$3s^23p^3\ ^2D^o_{5/2}$	(306 ± 10) ms	HSR [95]
Kr^{21+}	$3s^23p^3\ ^2D^o_{5/2}$	(0.80 ± 0.03) ms	EBIT [81]
S Sequence			
Ar^{2+}	$3s^23p^4\ ^1S_0$	(133 ± 24) ms	EST [85]
		$(159.7 \pm 9.7$–$38.4)$ ms	EST [67]
		(145 ± 5) ms	HSR [84]
Mn^{9+}	$3s^23p^4\ ^1S_0$	(21 ± 0.3) ms	EKT [78]
Mn^{9+}	$3s^23p^4\ ^1D_2$	(18.02 ± 0.16) ms	EKT [78]
Fe^{10+}	$3s^23p^4\ ^1D_2$	(9.86 ± 022) ms	EKT [37]
		(11.05 ± 0.1) ms	HSR [95]
Fe^{10+}	$3s^23p^3(^2D^o)3d\ ^3G^o_4$	(68 ± 4) ms	HSR [95]
Cl Sequence			
Mn^{8+}	$3s^23p^43d\ ^4F^o_{9/2}$	(210 ± 42) ms	EKT [80]
Fe^{9+}	$3s^23p^52P^o_{1/2}$	(13.64 ± 0.25) ms	EKT [37,79]
Fe^{9+}	$3s^23p^4(^3P)3d\ ^2F_{7/2}$	(17.0 ± 1.7) ms	HSR [95]
Fe^{9+}	$3s^23p^4(^1D)3d\ ^2F_{7/2}$	(4.9 ± 0.4) ms	HSR [95]
Fe^{9+}	$3s^23p^43d\ ^4F^o_{7/2}$	(85.7 ± 9.2) ms	EKT [80]
Fe^{9+}	$3s^23p^43d\ ^4F^o_{7/2}$	(93 ± 30) ms	EKT [80]
		(60 ± 10) ms	HSR [95]
Fe^{9+}	$3s^23p^43d\ ^2G^o_{9/2}$	(17.8 ± 3.1) ms	EKT [80]
Ar Sequence			
Kr^{18+}	$3s^23p^53d\ ^3P^o_2$	(2.20 ± 0.2) ms	EBIT [81]
Kr^{18+}	$3s^23p^53d\ ^3F^o_2$	(4.2 ± 0.5) ms	EBIT [81]
K Sequence			
Kr^{17+}	$3s^23p^63d\ ^2D_{5/2}$	(22.7 ± 1.0) ms	EBIT [81]
Ca Sequence			
Kr^{18+}	$3s^23p^63d\ ^{23}P_2$	(≈ 4.2) ms	EBIT [81]

EBIT Electron-beam ion trap, EKT ECR ion source plus Kingdon ion trap, HSR Heavy-ion storage ring, RFT Radiofrequency ion trap.

Kingdon trap [37,39,77–80]. Some of the measurements include 3d levels as well, some of which are long-lived, since no E1 decay channels are open. As the authors, Moehs and Church, state, the comparison of their experimental results (some claimed with errors as small as 0.9%, some with 30% uncertainty) with theory is inconclusive. However, very recent studies of similar levels by the storage ring technique (for an example, see Fig. 7) resulted in lifetime data that in various cases reached smaller uncertainties and did not necessarily confirm the Kingdon trap results. Thus part of the shortcoming may lie with some of the experiments. (Almost needless to say, being involved in the storage ring experiments with their better statistical reliability and more reliable measurement scheme, my confidence is more in my own results. However, the present ion injector at the TSR heavy-ion storage ring cannot supply ion currents of certain highly-charged ion species as high as necessary for some conclusive results.)

For many-electron ions beyond Fe ($Z = 26$), an EBIT may presently offer the best access for lifetime studies. For example, in a series of measurements on Si-like to Ca-like ions of Kr ($Z = 36$), two levels in Kr XIX (Ar-like) were observed that have major M2 decay branches [81].

Iron certainly is not the only element of astrophysical interest, although it is one in which some of the highest charge states of astrophysical interest (for example, Fe XIV/Fe^{13+}) occur. Forbidden transitions of astrophysical interest have been listed, for example, in papers by Eidelsberg, Crifo-Magnant and Zeippen [17] and by Lynch and Kafatos [18,19]. For a great many ionic systems with not too many electrons and with nuclear charges up to $Z = 42$ (Mo), forbidden transitions in the ground and some excited configurations have been compiled by Kaufman and Sugar [82]. Their work relies on calculations that are compared to the much fewer experimental wavelength data.

The differences between the experimental results, while often much larger than the combined error bars, are nevertheless much smaller than the declared uncertainties of

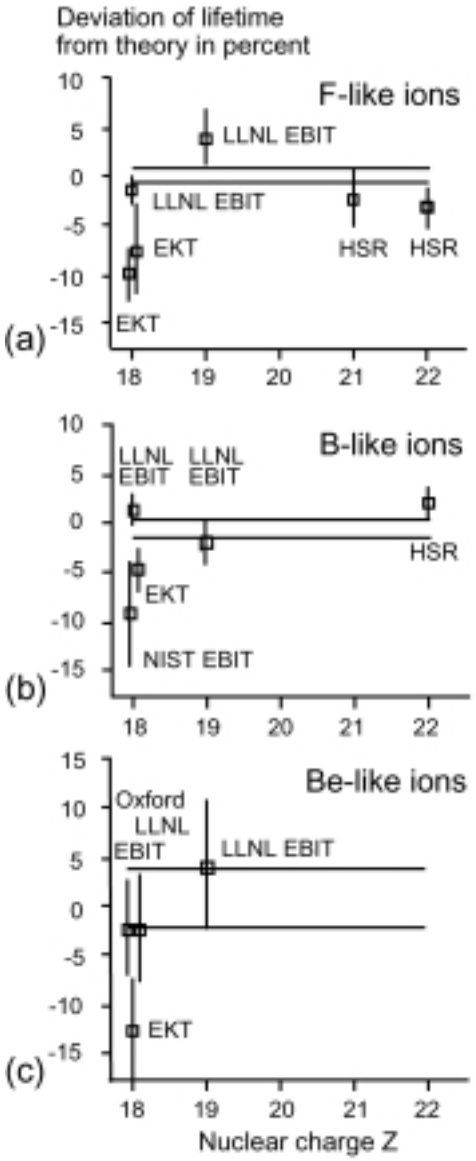

Fig. 6. Lifetime data for a) the $2s^2 2p^5\ ^2P^o_{1/2}$ level in the ground state of F-like ions, b) the $2s^2 p^2 P^o_{3/2}$ level in the ground state of B-like ions, and c) the $2s2p\ ^3P^o_2$ excited level of Be-like ions. In a) and b), all data are normalized to the theoretical results given by Cheng *et al.* [60] after semi-empirical corrections for experimental transition energies. In c), the data are normalized to the theoretical results given by Safronova *et al.* [74]. The horizontal bars indicate the scatter range of selected predictions (the predictive uncertainties are much larger). The experimental data are from an electrostatic Kingdon trap (EKT), from the NIST, Oxford and LLNL EBITs, and from a heavy-ion storage ring (HSR).

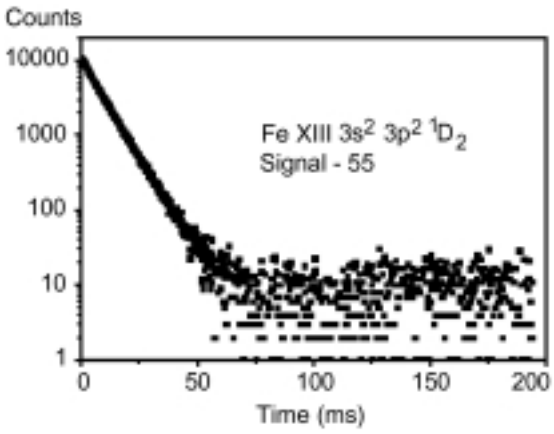

Fig. 7. Example of a decay curve recorded in the UV range with Fe^{12+} circling in the heavy-ion storage ring TSR [95].

most calculations (typically of the order of 10 to 20%), and that is much smaller again than the scatter of various predictions that may be only 20% in some cases and reach a factor of five in others.

In fact, many astrophysical observations have such poor statistical significance that for their interpretation transition rate reference data good to, say, 10% are sufficient so as not to contribute markedly to the error budget. The physics goal of more precise lifetime measurements is set much farther: Precise calculations for everyday atoms in our environment, that is mostly neutral particles and a few singly charged ions, are much more difficult than those for multiply-charged few-electron ions with a well-defined central potential. Also, extremely precise measurements as part of the quest for fundamental physics issues like parity violation require the study of heavy elements (because the non-classical effects are larger there), again a challenge for theory. Thus measurements on highly charged few-electron systems are one way to pursue quantum electrodynamical effects (for U^{91+}, theory seems ahead of experiment in terms of precision), but most other work on multi-charged ions is providing stepping stones in the quest for precise work on *neutral* and *near-neutral* atoms, for which nowadays relativistic calculations are being done despite their need for extreme computer resources. For example, measurements on low-charge state rare gas ions (Ne^{2+} [83] and Ar^{2+} [84] have been covered, Ar^{3+}, $Kr^{2+,3+}$ are wanted) ought to be done at a storage ring in order to check on the RF trap data [67,85,86] on such ions that are present in many technical plasmas. At TSR, these particular experiments require the use of a single-stage injector, since the rare gases do not possess the negative ions needed for the tandem accelerator that has been employed for most other ions.

4.5 *Hyperfine interaction*

So far I have presented spin change (intercombination) and higher-order multipole radiation (Ml, E2) as mechanisms behind the "slow" decay of long-lived excited levels. Let us now turn to a third process, transitions that are mediated by a coupling of the electron shell with nuclear momenta, that is by hyperfine interaction (hfs). The classical case is the ls2p $^3P^o_0$ level in He-like ions that normally cannot decay to the $1s^2\ ^1S_0$ ground state because absolutely no single-photon 0-0 transitions are allowed. However, with a non-vanishing nuclear spin, there may be hyperfine components that have the same total angular momentum quantum number $F = J$ (electron shell) + I (nucleus). Then a hyperfine structure sublevel of the $^3P^o_0$ level mixes with a hyperfine structure sublevel of the $^3P^o_1$ level, which in turn mixes with the $^1P^o_1$ level (multiplet mixing). Obviously, the $^3P^o_0$ level is much longer lived than its partner level $^3P^o_1$, which in turn is longer lived than the $^1P^o_1$ resonance level. Beam-foil spectroscopy has demonstrated and used this effect in various He-like ions, for example to obtain information on fine structure intervals that were not spectroscopically resolved (for references, see [46]). If the K-shell is filled however, the lifetimes of the resonance levels with which hyperfine interaction mixes the otherwise long-lived levels are not quite as short as in He-like ions. Consequently, the hfs-induced lifetimes of interest in Be- and Mg-like ions will be in the

millisecond and microsecond ranges [87,88] – clearly a topic for future electron beam ion trap studies.

Let us now turn to a truly fundamental case, a heavy ion with only a single electron, like $^{209}Bi^{82+}$. The prototypical one-electron system, hydrogen, has a nuclear spin, and thus the ground state features hyperfine splitting. The spin-flip between the two hyperfine levels gives rise to the 21-cm line of radioastronomy and to the hydrogen maser radiation. Because of Z^3 scaling, the same transition in hydrogen-like $^{209}Bi^{82+}$ is in the near ultraviolet. This has been confirmed by a laser-resonance experiment on H-like Bi ions circling in the heavy-ion storage ring ESR at GSI Darmstadt [25]. The search for the transition was tedious, because the actual value of the transition energy differed somewhat from expectation. This then lead to the need for improving the nuclear model, for example assuming a "soft" distribution of the nuclear magnetic moment. Similar corrections were necessary for the interpretation of wavelength data that were obtained using the Livermore EBIT on isotopes of Re, Ho and, most recently, TI [89]. The unique advantage of the storage ring, however, is the capability for lifetime measurements, which is here effected by pulsed laser excitation. The latest of these measurements is impressively precise (0.4%) [27]. Meanwhile, studies have covered a second isotope, $^{207}Pb^{81+}$, that required additional experimental tricks [26]. The lifetime value extracted here is not (yet) as precise as the above.

5. Prospects

The electron beam ion trap (EBIT) has the enormous advantage of being applicable to many elements, and in particular to high charge states. EBITs will be essential to identify various forbidden lines that appear in the VUV and EUV spectrum of the solar corona. EBITs can be used to establish the elemental species and the ionization energy of the ions that emit these lines, and it is (fairly) straightforward to obtain decay curves, too for levels in the lifetime range from a fraction of a millisecond to many milliseconds, with error margins of a few percent for observations in the visible spectrum (limited because of not yet understood systematic influences) and much better than that in the X-ray range.

The heavy-ion storage ring is the most promising tool for precision lifetime measurements, because it combines isotope and charge state selectivity with very good vacuum conditions and easy control of the stored ions and their storage behaviour. Lifetime error margins of about 0.2% have been reached by several experimental techniques at TSR, and further improvements in precision are quite feasible, though not exactly cheap in terms of equipment and accelerator time.

Such a high precision of atomic transition rate data seemed out of reach only a few years ago when astrophysics demanded better atomic data in order to sensibly interpret the data that were streaming in from various new telescopes. Now some laboratory data are available that exceed the immediate quality needs of the astrophysical data evaluators, and the question arises whether to produce many more "sufficiently good" or a few "better" lifetime data. Of course, lifetimes alone are not always sufficient (but many of the above examples are of unbranched decays, permitting a direct conversion of lifetimes into transition rates). Good branching ratios are also needed to determine individual transition rates in branched decays and to properly assess the weaker transitions which do not contribute much to the lifetimes.

However, benchmark data are required to test and "calibrate" calculations that are needed to treat the many cases not measured, in particular the codes and data bases used for collisional-radiative models employed in large-scale syntheses of solar and stellar spectra and in laboratory plasma diagnostics. Under closer scrutiny, even purportedly "complete" models are demonstrably incomplete and error-ridden as concerns the atomic level structure, and systematic tests of the transition rates employed may not even have been done. Anyway, why not – as the means are there – leapfrog from the previous state of missing sufficiently meaningful experimental lifetime data on certain cases to offering precise data that challenge the present calculational approaches? Electron beam ion traps and heavy-ion storage rings can do it.

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